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Environmental signals in limestone-marl alternations**

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### Environmental signals in limestone-marl alternations

Florian Böhm<sup>1</sup>, Hildegard Westphal<sup>2</sup>, Stefan Bornholdt<sup>3,4</sup>

- 1) GEOMAR Forschungszentrum, Wischhofstr. 1-3, 24148 Kiel, Germany, fboehm@geomar.de;
- 2) Institut für Geologie und Paläontologie, Universität Hannover, Callinstr. 33, 30167 Hannover, Germany, hwestphal@geowi.uni-hannover.de;
- 3) Institut für Theoretische Physik, Leibnitzstr. 15, 24098 Kiel, Germany
- 4) current address: Interdisziplinäres Zentrum für Bioinformatik, Universität Leipzig, Kreuzstr. 7b, 04103 Leipzig, Germany, bornholdt@izbi.uni-leipzig.de

#### Abstract

The nature of rhythmic carbonate-rich successions such as limestone-marl alternations has been, and still is, subject to controversy. The possibility of an entirely diagenetic origin for the rhythmic calcareous alternations is discarded by most authors. One problem with an entirely diagenetic, self-organized development of limestone-marl alternations is the fact that limestone and marl beds in many examples are laterally continuous over several hundred metres or even kilometres. In an entirely self-organized system, lateral coupling would be very limited; thus one would expect that, rather than laterally continuous beds, randomly distributed elongate nodules would form.

We address the origin of limestone-marl alternations using a computer model that simulates differential diagenesis of rhythmic calcareous successions. The setup uses a cellular automaton model to test if laterally extensive, rhythmic calcareous alternations could develop from homogeneous sediments in a process of self-organization. Our model is a strong simplification of early diagenesis in fine-grained, partly calcareous sediments. It includes the relevant key mechanisms to the question of whether an external trigger is required in order to obtain laterally extensive limestone-marl alternations.

Our model shows that diagenetic self-organization alone is not sufficient to produce laterally extensive, correlatable beds. Although an external control on bedding formation could be considered to have solved the problem as commonly assumed, we here suggest

an interesting third possibility: the rhythmic alternations were formed through the interaction of both an external trigger and diagenetic self-organization. In particular we observe that a very limited external trigger, either in time or amplitude, readily forms correlatable beds in our otherwise diagenetic model. Remarkably, the resulting rhythmites often do not mirror the external trigger in a one-to-one fashion and may differ in phase, frequency and number of couplets. The interpretation of calcareous rhythmites as a one-to-one archive of climate fluctuations therefore might be misleading. Parameters independent of diagenetic alteration should be considered for unequivocal interpretation.

*Keywords:* Carbonate diagenesis; climate archives; limestone-marl alternations; self-organization; cellular automaton; computer simulation

## 1. Introduction

The origin of calcareous rhythmites such as limestone-marl and limestone-shale alternations has been subject to controversy for several decades (see, e.g., Bathurst, 1971; Einsele, 1982; Milliman, 1974; Einsele et al., 1991). Especially controversial is the question of diagenetic alterations overprinting or causing the rhythmic nature of these successions. While for some successions the primary, sedimentary nature of the cyclicity is clearly proven by differences between the fossil content of limestones and marls (e.g., Weedon and Jenkyns, 1999), it is surprisingly difficult to prove a sedimentary origin for other successions (Sujkowski, 1958; Eder, 1982; Hallam, 1986; Munnecke and Samtleben, 1996; Westphal et al., 2000). Contributing to the problem is the fact that the primary sediment composition (fossil content as well as mineralogical composition) can be disguised during diagenesis by selective dissolution (e.g., of aragonitic components; Dullo, 1990; Bathurst 1987) and by passive enrichment. Therefore, the differences in carbonate content, sedimentary components or trace elements in diagenetically mature rhythmites do not necessarily reflect primary differences in the sedimentary succession (Bathurst, 1987).

Since the work of Ricken (1986) it is generally accepted that diagenesis strongly enhances differences in the initial sediment composition. The interpretation of rhythmites as reflecting diagenetically enhanced primary fluctuations is dominant in the current literature. Such fluctuations in carbonate supply have been termed productivity cycles (Seibold, 1952), whereas periodic increases in the clay supply are known as dilution cycles (e.g., Einsele, 1982). Nevertheless, several successions have been described that lack indications of systematic initial variations in the sedimentary composition. For such successions it has been proposed that they were produced merely by diagenesis. A purely diagenetic origin of rhythmic alternations from homogeneous precursor sediment has been interpreted, e.g., for Devonian and Carboniferous limestone-marl alternations from Germany (Eder, 1982) or for Silurian limestone-marl alternations from Gotland (Munnecke, 1997; Munnecke and Samtleben, 1996). In the latter case, it was shown that the only difference in fossil associations between limestones and marls is the systematic absence of aragonitic components in the marls. This, however, is not interpreted as an original sedimentary signal, but as a diagenetic overprint by selective dissolution of aragonite in what became marl layers (Munnecke et al., 1997). This interpretation is corroborated by the findings of Cherns and Wright (2000) who studied the same formations and described preservation of aragonitic bioclasts in silicified sediment adjacent to marls without such aragonite components. This illustrates that faunal associations are biased by differential diagenesis.

Differential diagenesis in rhythmic calcareous successions has for a long time reminded geologists of another rhythmic phenomenon in the geological realm called “Liesegang rings”, which are secondary rhythmic colorations of sedimentary rock. Repetitive mechanisms similar to those causing Liesegang rings are common in present-day geochemical systems out of equilibrium. Repetitive processes can result, for example, in repeated precipitation and replacement (Liesegang, 1913; Ortoleva, 1994; Ortoleva et

al., 1994; Ortoleva et al., 1993) or in magmatic crystallization with rhythmically varying trace element contents (Hoskin, 2000; Holten et al., 2000). Such processes are termed “self-organized”. Self-organization implies that the rhythmicity is a product of an inherent property of the system that is self-reinforcing and does not require any external cause. Similarly, Illies (1949) noted that in nodular limestones the distance between nodule-rich layers is remarkably constant and explained this observation by a self-organization process of dissolution and reprecipitation of carbonate particles during early diagenesis.

A problem with the interpretation of an entirely self-organized development of limestone-marl alternations is the fact that limestone and marl beds usually are laterally continuous over several hundred metres (Fig. 1) or even kilometres (Elrick and Read, 1991; Elrick et al. 1991). In an entirely self-organized system it is difficult to find a mechanism able to maintain a lateral coupling; therefore one would expect randomly distributed nodules rather than laterally continuous beds (Einsele, 1982). Ricken (1986) and Ricken and Eder (1991) assumed that minor initial differences in the precursor sediment are enhanced by diagenesis. Such minor fluctuations could possibly act as a lateral coupling mechanism. As a result of the thorough diagenetic reorganization of carbonate sediments, however, such minimal sedimentary differences would be difficult or even impossible to prove in many cases.

The present study uses a simple cellular automaton model to test if laterally extensive, rhythmic alternations could in principle develop from homogeneous sediments in a process of self-organization. We further examine the rôle of an external trigger in such a system and whether its imprint is itself modulated through interferences with self-organized diagenetic mechanisms.

## **2. Geological Background**

Our numerical model is a strong simplification of early diagenesis in fine-grained calcareous sediments. It includes the relevant key mechanisms to answer the question as to whether an external trigger is required in order to get laterally correlatable, extensive limestone–marl alternations. Our algorithm is based on the diagenetic model of Eder (1982) as modified by Munnecke and Samtleben (1996) and Munnecke (1997). Following Munnecke and Samtleben (1996) and Munnecke (1997) our numerical model employs metastable aragonite as the “engine” of rhythmic diagenesis.

Recent studies of initially aragonite-bearing calcareous successions have shown that lithification of diagenetically metastable carbonate sediments occurs during early marine burial diagenesis (Melim et al., 1995; Melim et al., 2001; Munnecke, 1997; Munnecke et al., 1997; Westphal, 1998). As these authors have shown, diagenetically mature limestones with tight calcite cement framework are lithified during early marine burial diagenesis. Lithification takes place before mechanical compaction could lead to the deformation of burrows and fossil tests. In rhythmites altered during early marine diagenesis, interlayers between such limestone beds are devoid of a calcite cement framework, and in contrast to the limestone beds, they are always strongly mechanically

compacted. These interlayers are interpreted as donor beds where aragonite has been dissolved to be reprecipitated in the limestone beds as calcite cement (Westphal et al., 2000; Munnecke et al., 1997).

This model of early differential diagenesis of initially aragonite-rich sediments, with aragonite dissolution in the marls and reprecipitation of the dissolved calcium carbonate as calcite cement, is in contrast to diagenesis of pure calcitic sediments that are mainly altered by pressure dissolution and reprecipitation during deep burial diagenesis (Borre and Fabricius, 1998).

A three-step diagenetic process is postulated for the diagenesis model after Eder (1982) as modified by Munnecke and Samtleben (1996) (Fig. 2). **(1)** After sedimentation, the aragonite-bearing sediment layer A is slowly buried and passes through a stationary, layered, diagenetic environment. The sediment passes an aragonite dissolution zone, where aragonitic constituents are selectively dissolved. Calcite and insolubles are not affected. The dissolved calcium carbonate moves through the sediment column, and at least part of it follows the early compactional fluid expulsion and migrates upward. Above the aragonite dissolution zone, the dissolved calcium carbonate is reprecipitated as calcite cement in a sediment layer B. This cement drastically reduces permeability of this cemented sediment layer, thereby protecting it against dissolution during its later migration through the aragonite dissolution zone. Early cementation and resulting mechanical stabilization at shallow burial depth are indicated by the uncompacted or only slightly compacted nature of the limestone layers (Munnecke et al., 1997). The zone of aragonite dissolution and cement reprecipitation is thought to be located within a few metres below the sediment-water interface and presumably is the result of bacterial activity (see Canfield and Raiswell, 1991). **(2)** When this cemented layer B enters the dissolution zone, less aragonite is dissolved and consequently less dissolved calcium carbonate is available for cementing the layer C above. **(3)** Upon entering the aragonite dissolution zone, this uncemented layer C shows again less resistance against aragonite dissolution because no cementation is sealing it. This model potentially leads to a self-organized oscillation between carbonate receiver and carbonate donor beds (limestones and marls), even when starting from a homogeneous sediment.

A slightly different model for self-organized diagenetic generation of limestone layers was developed by Jenkyns (1974) for Jurassic red nodular limestones of the Mediterranean region (Fig. 3). Jenkyns (1974) also assumed metastable aragonite as the driving force for diagenetic alterations. The two models mainly differ in the dominant direction of pore-water flow that is driven by compaction and directed upward in the model of Munnecke and Samtleben (1996), and diffusion-driven downward in the model of Jenkyns (1974). Both models have in common that aragonite is being dissolved during early diagenesis and reprecipitated as low-Mg calcite cement. Jenkyns' three-step model includes the assumption of a downwards-directed pore-water movement. **(1)** Aragonite is dissolved in the sediment shortly after deposition while the sediment column is migrating through an aragonite dissolution zone. The dissolved calcium carbonate is reprecipitated below the zone of aragonite dissolution, where it forms layers of nodules. **(2)** With downward movement of the sedimentary column, the distance between the stationary

aragonite dissolution zone and the cemented nodules increases and finally becomes too large for transport of dissolved calcium carbonate. (3) The formation of new nodules then becomes energetically favourable and a new layer of nodules starts to form just below the aragonite dissolution zone.

In the model of Jenkyns (1974), the critical factor for the formation of distinct layers is the distance between the zone of dissolution and the preferred site of reprecipitation, whereas the critical factor in the model after Munnecke and Samtleben (1996) is the availability of aragonite for dissolution. Even though both models are based on different controlling mechanisms, they both theoretically result in rhythmic differential diagenesis that could lead to the formation of calcareous rhythmites such as limestone-marl alternations. In the following, the implementation of both models is described.

### 3. Methods

We use a cellular automaton to simulate the relevant key mechanisms of the two models described above, the model of Eder (1982), Munnecke and Samtleben (1996), and Munnecke (1997), and the model of Jenkyns (1974) (Fig. 4). The basic setup is identical for both models. Our automaton comprises a rectangular matrix of up to 500 active layers, each layer being 800 cells wide and an infinite number of inactive layers below, representing the sedimentary column. A cell is in one of three states (Fig. 5): (1) aragonitic, consisting of aragonite, calcite and clay minerals, (2) cemented, where calcite cement is added to the immature sediment, and (3) non-aragonitic, either originally aragonite-free sediment consisting of calcite and clay minerals, or aragonite-depleted diagenetically mature sediment consisting of calcite and clay minerals. Aragonitic (state 1) cells can be subject to aragonite dissolution, which transforms them into non-aragonitic (state 3) cells. Cementation, in contrast, converts aragonitic (state 1) and non-aragonitic cells (state 3) into cemented (state 2) cells. Layers of cemented and non-aragonitic (aragonite-depleted) cells form limestones and marls, respectively.

With each time step, the cells of a layer are moved one layer downward. Subsequently a new, uppermost layer with new sediment is added. The sediment in the new cells is either aragonitic (state 1) or non-aragonitic (state 3). Aragonitic and non-aragonitic cells are randomly distributed in the new layer. Their percentages are predefined and may vary systematically between layers to form an external trigger (see below). An overview of all adjustable parameters of the model is given in Appendix 1.

A dissolution zone of specified thickness of one to several layers is located in a specified depth. As a layer passes through the dissolution zone, aragonite is dissolved from state (1) cells. The cells in the dissolution zone are checked in random order for dissolvable aragonite. For each cell from which aragonite is dissolved, a specified number of cells in the cementation zone is instantly cemented. The cementation zone is situated at a specified distance from the aragonite dissolution zone. For the model after Munnecke and Samtleben (1996), the cementation zone is located above the aragonite dissolution zone, for the model after Jenkyns (1974), the cementation zone is located below the

dissolution zone. Both, aragonitic and non-aragonitic cells can be cemented in the cementation zone. If there is no cell available for cementation, the dissolved carbonate is lost from the system.

For the model of Munnecke and Samtleben (1996), a layer A altered in the cementation zone will enter the dissolution zone after a number of time steps, depending on the distance between cementation zone and dissolution zone. In addition to cells that have been already cemented in the cementation zone above (state 2), layer A may contain pristine aragonitic cells (state 1). In the dissolution zone, this aragonite will dissolve in state 1, pristine aragonitic cells, whereas cemented state 2 cells are assumed to be effectively sealed from aragonite dissolution, a key mechanism of the model. Once layer A has passed below the dissolution zone, the state of its cells remains unchanged.

By way of contrast, in the model of Jenkyns (1974), where the cementation zone is located below the dissolution zone, a layer A enters the dissolution zone without previous cementation. All aragonite of state 1 cells is subject to dissolution. Cementation takes place below the dissolution zone. Once the layer A has passed below the cementation zone, the cells are not subjected to further change.

The depth and thickness of dissolution and cementation zones are defined by the modeller. In the simplest case, both zones are only one layer thick. A greater thickness leads to a more fuzzy distribution of cemented/uncemented cells in the diagenetically mature sediment. If the dissolution zone has a thickness of more than one layer, it is represented as a row of columns rather than cells. In this case, from each column of the dissolution zone a cell is randomly selected at every time step and aragonite is dissolved from it if it is aragonitic (state 1). This random selection may be repeated for a specified number of times during each time step.

In the simplest case the cell to be cemented is positioned vertically above the cell from which the carbonate was dissolved. If the modeller defines that more than one cell is cemented by the aragonite dissolved from one cell, the cementation procedure described below is repeated appropriately. Moreover, to allow for some horizontal transport of the dissolved calcium carbonate, a cementation matrix with specified width and thickness can be defined, centered above the cell, where aragonite is being dissolved. Within this matrix a cell is picked randomly and tested if it can be cemented (i.e. if it is not cemented yet). The probability for each cell to be picked within the cementation matrix can be predefined. This allows, for example, the possibility of assigning a higher cementation probability for cells directly above the dissolved cell. After a cell is picked for cementation, it is checked for cemented neighbours. Cementation of a cell next to an already cemented cell is preferred to account for the mechanism of accretion found in nature. If no cemented neighbours are present to attach to, a new seeding is necessary. Formation of new seed crystals occurs with a small predefined probability. If no new seed is formed or if the chosen field is already cemented, the search is repeated for a specified number of trials.

To check if the cell chosen for cementation is a cell that can be cemented with a low expense of energy, i.e., a cell with many already cemented neighbours, the algorithm now allows for a migration of the dissolved calcium carbonate for a limited, specified distance

to find a cementation site with more cemented neighbours (Fig. 6). This is intended to simulate the affinity of cementation to form compact patches, i.e., concretions. The search direction is selected randomly. The dissolved calcium carbonate can only move to cells with more cemented neighbours than the previous cell. Cemented cells block the flow.

A variation in the initial aragonite content of a pristine, newly deposited layer is used to implement an external trigger, i.e. a sedimentary signal. In nature, variations in aragonite can result, e.g., from sea-level fluctuations (Droxler et al., 1983). We use a simple switch between two states of either high or low percentages of aragonitic cells. Sedimentation switches from one state to the other either with a regular periodicity (periodic trigger) or at random intervals (random trigger). In the latter case an upper and lower boundary for the duration of these intervals is specified. This latter approach simulates a quasiperiodic switch with the width of the spectral power distribution depending on the specified boundaries.

In all sequential operations, cells are accessed in random order to avoid artificial anisotropies. For generation of random numbers we use the "ran3" and "irbit2" algorithms of Press et al. (1989). Pascal source code is available from the authors. Frequency analyses of the simulation results were carried out using the SSA-MTM toolkit of Ghil et al. (2002).

## 4. Simulation runs

### 4.1 Runs without external trigger

In the simplest case, we run the model with a purely deterministic behaviour. In this case all random, noise-generating processes are turned off and the simulation starts with a homogeneous sediment consisting entirely of aragonitic cells (state 1 cells; Tab. 1). At start-up, aragonite is available in all cells and dissolves synchronously in all cells entering the dissolution zone, which leads to the formation of a distinct cemented layer in the cementation zone. For the model of Munnecke and Samtleben (1996), where the cementation zone is situated above the dissolution zone, the model produces a vertical succession of distinct cemented and uncemented layers (Fig. 7a). The thickness of these layers is determined by the vertical distance between the centre of the cementation zone and the centre of the dissolution zone. Consequently, power spectra of the cement density distribution along the succession show a main peak at a periodicity of twice the distance between dissolution layer and cementation layer. Additionally, there are harmonics at  $1/3$  and  $1/5$  of this wavelength that are an effect of the non-sinusoidal shape of the diagenetic signal. However, laterally continuous layers are only formed, because the simulation start synchronizes the top layer cells. The initial boundary conditions are required for the formation of continuous layers, because there is no implicit synchronization mechanism in the deterministic case of the model. It merely repeats the initial conditions *ad infinitum*. If the model is started with an initial random distribution of cemented nodules, no layers are formed but the random nodular pattern is repeated again and again (c.f. Fig. 7e).

To get a more realistic representation of natural processes we included several noise processes. We attributed a finite thickness to the dissolution and the cementation layers. A random search algorithm selects cells for dissolution or cementation from the columns of cells defined by this thickness. We also allow for a limited horizontal movement of solutions. For this purpose we use a cementation matrix of a designated height and width. Cementation is further controlled by a limited probability to generate new crystal seeds, resulting in agglomeration of cemented cells. Finally, only part of a newly formed layer is filled with dissolvable aragonitic cells, which are randomly distributed. This results in a lower amount of cement per layer dissolved.

The widening of the cementation layer results in a dissemination of cemented cells, especially if sufficient crystal seeds are available (high seeding probability; Tab. 1). No sharply defined cemented layer is formed, but a diffusely cemented zone (Fig. 7b). Lower seeding probabilities lead to either dendritic or nodular cementation patterns. With a low seeding probability, a relatively thin cementation layer promotes vertical aggregation of cement, as time for horizontal growth is limited. The formation rates of new seeds can be further reduced if dissolved aragonite is allowed to move horizontally, i.e., with a wide horizontal range for fluid migration. This is because with a wider horizontal range, fluids from dissolved cells have a higher probability to find existing cements to attach to and thus do not need to form new seeds. Therefore, if a thin cementation layer is combined with a very low seeding probability and a wide horizontal range for fluid migration, the model generates dendritic structures (Fig. 7c, Tab. 1). This behaviour is expected due to the diffusion-limited character of the aggregation mechanism in this special case. A more realistic representation of cementation processes is achieved, if a mechanism is included that favours attachment of new-cemented cells not only to a preformed seed but also to sites with maximum numbers of cemented neighbour cells. This mimics the energetic benefit of a minimal surface to volume ratio. With such a mechanism our model produces a nodular or patchy cementation pattern if the cementation layer is sufficiently thick that nodules can grow in width and not only in height (Fig. 7d, Tab. 1).

If we now position the cementation zone below the dissolution zone, our cellular automaton operates similar to the model of nodule formation suggested by Jenkyns (1974). We restrict dissolution to the top layer. Dissolved aragonite is allowed to diffuse downwards to form calcite cements as suggested by Jenkyns (1974). A decreasing cementation probability from top to bottom of a thick cementation layer mimics a diffusion gradient. Formation of new seeds therefore becomes more likely relative to the aggregation to existing cements, the deeper the dissolved aragonite has to move. In this mode, our simulation produces cemented nodules as expected by Jenkyns (1974). However, no mechanism is available to synchronize the seed formation and nodule growth. Thus, unlike the expectations of Jenkyns (1974), our model shows that nodules are not arranged in layers but rather are randomly distributed (Fig. 7d). The mean size of the nodules formed in this way is controlled by the seeding probability. Lower probabilities lead to larger nodules. The thickness of the cementation layer, on the other hand, has no significant influence on nodule size, but rather controls nodule shape. As for the model after Munnecke and Samtleben (1996), a thin cementation layer promotes

attachment of cement to the top of growing nodules and may lead to columnar or dendritic shapes.

For the model after Munnecke and Samtleben (1996), the concentration of cemented cells to a densely cemented, confined layer is the major control on the formation of layering versus diffuse or nodular cementation. As a layer reaches the dissolution zone, a high concentration of cemented cells, and consequently low concentration of pristine aragonitic cells, results in a low availability of dissolved calcium carbonate to act as a source of cement. This keeps the diagenesis oscillator in its no-cementation mode. The concentration of aragonitic cells in newly formed layers and the thickness of the cementation layer are primary control factors. The thinner the cementation layer, the more cemented cells are concentrated to a confined narrow zone to form a solid bed. This counteracts dissemination of cemented cells. The same is true for a high percentage of aragonitic cells. If aragonite concentration is increased continuously during a simulation, starting with a low aragonite percentage level, the formation of more or less continuous, cemented layers or lenses starts after a certain threshold density is reached (Fig. 7e, Tab. 1). However, lateral synchronization of this layering is only brought about by the synchronous aragonite increase of the newly sedimented layers. If this increase is slow (as in Fig. 7e), no synchronization is achieved. Furthermore, small random disturbances of the distribution of cemented cells will propagate through the stack of layers and disrupt a well-defined layer structure within a short distance (Fig. 7f). Therefore, without an external trigger, the two models studied here fail to generate or even maintain a laterally continuous layering, when realistic conditions, including random disturbances, are included.

#### *4.2 Influence of external triggers*

In a second step we investigated the influence of an external trigger signal on the diagenetic partitioning of cemented and uncemented layers. As an external signal we used step-like changes in the proportion of dissolvable aragonite in the pristine sediment. In the previous chapter it was shown that the initial conditions of a simulation run can synchronize cementation and lead to the formation of several isochronous cemented layers. However, in simulations with a realistic representation of noise processes in the diagenetic environment such layering quickly disintegrates (Fig. 7f, Tab. 1).

We observed that this disintegration is counterbalanced by including an external signal in the simulation, that results in continuous layer formation even in the presence of noise (Figs. 8a, 9a, Tab. 2). The external signal, though, is not recorded in the rhythmic succession in a simple one-to-one fashion. Interactions between trigger signals, i.e. the sedimentary response to the environmental forcing, and cementation cycles lead to complex bedding patterns. The following phenomena were observed:

(i) Bundling of cemented beds occurs if the recurrence time of the trigger signal is much longer than the periodicity of the diagenetic cycles (Fig. 8a, Tab. 2). The number of

resulting couplets is larger than the number of trigger signals, and both frequencies, of the trigger and the diagenetic system, are expressed in the resulting succession (Fig. 10a).

(ii) An external trigger signal, even if random or quasi-periodic, can maintain regular cyclic diagenetic bedding (Fig. 8b, Tab. 2). However, couplets of cemented and uncemented beds do not correspond to the external trigger in a one-to-one fashion in the resulting succession. The position of the cemented beds in the sedimentary column may shift with respect to the original position of the trigger event layer; that is, the cemented bed is located above the corresponding aragonite-rich bed (Fig. 9c). A periodogram of the time series of the external trigger and the time series of cement content in the diagenetically altered succession reveals that both have a common broad peak centered at the mean trigger wavelength. However, in the time series of the diagenetic cement content, additional peaks are present, that lack a counterpart in the time series of the trigger signal (Fig. 10a). These higher frequency periodicities in cement content are a purely diagenetic signal and do not carry any palaeoenvironmental information.

(iii) In the case of similar frequencies of trigger signal and diagenetic oscillator, the resulting succession apparently records the external signal. However, a phase shift of the diagenetic beds may occur, even towards a phase locking  $180^\circ$  off the trigger signal as illustrated in Figure 9b.

(iv) If the average periodicity of the trigger is equal to or shorter than the wavelength of the diagenetic bedding, the relationship between trigger signal and diagenetic bedding gets even more complex (Fig. 8c). This situation would result, for example, from slow sedimentation. The phenomena observed in cases i-iii occur in such a setting as well; i.e., cemented beds do not coincide with initially aragonite-rich layers and are shifted vertically with respect to these layers; the number of cemented beds may differ from the number of initially aragonite-rich layers. Additionally, in frequency analyses, the cement content spectra may entirely lack a peak corresponding to the trigger period. This is demonstrated by the six experiments shown in Fig. 8c. An identical sequence of trigger pulses was used for these experiments; only the thickness of the layer between the cementation zone and the dissolution zone varies (Tab. 2). The formation of the first bed in each experiment is controlled by the initial conditions, i. e. by the time it takes the first aragonite-rich layer to reach the dissolution layer. This start-up phase is reflected in the cement-free zone at the base of each column (Fig. 8c). The formation of the following beds, however, is controlled by the trigger sequence and its interaction with the diagenetic cyclicity. Frequency spectra illustrate that the diagenetic bedding frequencies differ in these six simulations (Fig. 10b). In some cases the trigger signal is not preserved at all. This means that the periodicity of the sedimentary succession may not preserve any information at all about the external, environmental signal that keeps up the diagenetic bedding cycles.

## 5. Discussion and Implications

Our computer-based diagenesis simulation is the result of an implementation of diagenesis models based on geological observations. It represents a simplification of real-world processes and has to be understood as such. Nevertheless our model points out that there are potential pitfalls in interpreting limestone-marl and limestone-shale alternations. Our simulations agree with the widely accepted notion that diagenetic self-organization alone is not sufficient to result in laterally extensive, correlatable beds (see Einsele, 1982). The continuous beds seen in our simulation runs without an external trigger are the result of the initial boundary conditions of the model and are lost after some simulation time. In our simulations, the absence of external lateral correlation mechanisms cannot be replaced by enforcing lateral accretion of diagenetic nodules. Without an external signal, no isochronous beds are formed.

Including an external trigger we find that this problem of lateral correlation is solved, as commonly assumed; however, we observe that this does not need to imply synchronicity with the external signal. Rhythmic alternations could also form as a complex interaction between an external trigger and the diagenetic dynamics. An external trigger readily induces formation of laterally correlatable beds in our otherwise diagenetic model. However, the simulated rhythmites in many cases do not reflect the external trigger in a one-to-one fashion. They may differ in phase, frequency and number of couplets. Rhythmic alternations may show higher frequency than the external signal, such as sub-Milankovitch cyclicity. Also, in contrast to intuition, cementation does not necessarily take place in the aragonite rich layer that initiates the differential diagenesis process. A vertical shift might occur between the originally aragonite-rich layer, where aragonite dissolution takes place, and the cemented layer, the distance of which depends on the distance of transport of the dissolved calcium carbonate.

Most striking in terms of the interpretation of limestone-marl/shale alternations are the observations made on the six simulations with identical trigger (Fig. 8c). This multitude of sedimentary successions is produced by one and the same trigger signal simply through interaction with diagenetic cyclicities of different wavelengths. A geologist attempting to correlate these sedimentary successions in a bed-to-bed correlation would find traceable marker beds, some of which can even be followed through all six columns. However, none of these marker beds is isochronous in the six columns that were simulated using identical constant sedimentation rates, i.e., the vertical axis is a linear representation of time (Fig. 8c). Furthermore, many beds are present only in one column or split into two beds in an adjacent column. Finally, most of these presumed correlations are caused by the start-up conditions and correlative beds become rare in the upper part of the columns.

This implies that the records of rhythms found in nature do not necessarily have significance in terms of climate or other rhythmic changes of the environment that are thought to cause such alternations. Curiously, in certain rhythmites neither calcareous components nor the diagenetically more inert palynomorphs show systematic variations between cemented and uncemented beds that would support a primary environmental signal as a cause for the rhythmic appearance (Westphal et al., 2000). This means that direct interpretation of rhythmites as recording climatic signals could be misleading and should be undertaken cautiously. The inherent enigma in determining the primary versus

diagenetic nature of limestone-marl or limestone-shale rhythmites is that numerous commonly studied parameters are not or not entirely resistant to diagenetic alterations. This is obviously true for carbonate, clay and organic matter contents, but also for petrophysical parameters such as natural gamma ray and porosity. Another parameter frequently examined is associations of calcareous fossils. However, these associations can also be considerably modified during diagenesis by selective dissolution (diagenetic sieve, Dullo, 1990; Bathurst 1987). For determining the nature of calcareous rhythmites, parameters are needed that are independent of the diagenetic system. There are surprisingly few such parameters, as for example the associations (not the concentrations!) of diagenetically relatively inert palynomorphs (compare, e.g. Westphal et al., 2000). Without such independent parameters, a conclusion as to whether one is looking at climatically-steered rhythms or climatically-meaningless diagenetic cycles must be equivocal.

## **6. Conclusions**

Diagenetic self-organization alone is not sufficient to result in laterally extensive, correlatable beds. Our model suggests that some external trigger, such as changes in sediment composition, initial porosity, or organic content, is a necessary prerequisite for the generation of laterally continuous, correlative limestone-marl and limestone-shale couplets. Additionally, we observe that in our simulations rhythmic alternations form as an interaction of both, an external trigger and diagenetic self-organization, reducing the role of the external trigger to inducing lateral correlation, whereas temporal synchronization between the sediment alternations and the trigger's time sequence may be altered or even lost in this process. This means that the rhythmites in many cases do not mirror the external trigger in a one-to-one fashion. They may differ in phase, frequency and number of couplets.

Time series of rhythmites resulting from an external trigger and diagenetic self-organization could exhibit clear periodic signals that can be confused with periodicities of orbital forcing. This implies that direct one-to-one interpretation of rhythmites as recordings of climatic signals should be undertaken cautiously, preferably including diagenesis-independent parameters.

## **Acknowledgements**

We are thankful to Axel Munnecke for many fruitful discussions. This research was partly supported by the German Science Foundation (DFG project WE 2492-1). Reviews of M. Tucker, D. Granjeon, and W. Schwarzacher considerably improved the manuscript and are gratefully acknowledged.

## Appendix 1

Simulation runs of the cellular automaton model are controlled by the adjustable parameters listed below. The complete Pascal source code listing is available from the authors.

*AragPerc1*: percentage of aragonitic cells in new sediment layer. Above a threshold value of about 0.35-0.45 cementation becomes sufficiently dense to cause self-organized separation of cemented and uncemented layers. Model sensitivity to this parameter is highest if AragPerc is in this range of values.

*AragPerc2*: alternates with AragPerc1 in specified or random intervals to simulate an external trigger signal. The model is sensitive to the difference between AragPerc1 and AragPerc2. High amplitude variations of these two parameters exert a strong control on cementation density.

*cdratio*: number of cemented cells per dissolved cell. Acts as amplifier of the aragonite percentage specified by AragPerc and therefore has a strong control on the simulation runs, namely the cementation density.

*SeedProb*: Probability of formation of new seeds for cementation. Mainly controls the distribution and shapes of cemented patches when cementation density is relatively low. Important control on the size of cement nodules.

*CemThick*: thickness of the cementation layer. Controls the concentration of cemented cells to a confined layer. A thick cementation layer leads to more diffuse cementation and weakens the self-organized separation of cemented and uncemented layers. A very thin cementation layer causes the growth of columnar or dendritic structures instead of nodules.

*CemWidth*: width of the cementation matrix, a matrix of CemWidth x CemThick cells from which a cell for cementation is selected. Large values allow some horizontal movement of the dissolved carbonate before forming a cemented cell. Simulations are not very sensitive to changes of this parameter.

*SearchMax*: number of searches for cementation site with maximum of cemented neighbour cells. Higher values result in higher cementation density. Simulations are not very sensitive to changes of this parameter.

*CemMax*: number of trials to find a cell for cementation in the cementation matrix. Low values lead to less efficient cementation and more loss of dissolved carbonate. Simulations are not very sensitive to changes of this parameter.

*DissThick*: thickness of the dissolution layer. A thick dissolution layer leads to slightly more diffuse cemented layers. At very low values aragonitic cells may pass the dissolution zone undissolved. Simulations are not very sensitive to changes of this parameter.

*DissMax*: number of trials to find a cell for dissolution in the dissolution layer. If the selected cell is already dissolved or is not dissolvable (state 2 or state 3 cells), another cell may be selected. Simulations are not very sensitive to changes of this parameter.

*DissTop*: position of the top of the dissolution layer. The distance between the centre of the dissolution layer and the centre of the cementation layer controls the thickness of cemented and uncemented layers.

*CemTop*: position of the top of the cementation layer. A position of the cementation zone below the dissolution zone allows the model to operate similar to the model of nodule generation of Jenkyns (1974).

*CycMax*: duration of a trigger half-cycle. *AragPerc1* is valid for this period after which *AragPerc2* is valid for the second half of a cycle. If a simulation works with a quasiperiodic trigger cycle, this parameter defines the maximum duration of a trigger half-cycle. Trigger cycle duration may have a strong influence on the simulations in certain cases discussed in the text.

*CycMin*: minimum duration of a trigger half-cycle for the quasiperiodic trigger cycle mode. Difference between *CycMin* and *CycMax* defines the frequency range of the trigger cycles.

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## Figure Captions

Fig. 1: Typical limestone-marl rhythmite with laterally extensive beds. Limestone layers are more resistant to weathering than marl layers, resulting in the characteristic layered aspect of such rhythmites in outcrop. Locality Hörnlebruch, Upper Jurassic, Southwest Germany.

Fig. 2: Simplified schematic diagram of the diagenesis model for rhythmic calcareous successions of Munnecke and Samtleben (1996) and Munnecke (1997) illustrating the diagenetic redistribution of carbonate, aragonite dissolution/calcite precipitation (compare Eder, 1982). (I) An aragonite-bearing sediment enters the shallow-burial diagenetic realm. Aragonite is dissolved in the aragonite dissolution zone and migrates diffusively to an overlying zone of calcite reprecipitation. (II) When a previously cemented layer enters the aragonite dissolution zone, the aragonite constituents are protected from dissolution by the surrounding calcite cement. During this time increment, aragonite dissolution is drastically reduced, and consequently only minor calcite cement reprecipitation takes place. (III) When this uncemented layer enters the aragonite dissolution zone, the unprotected aragonite is subject to dissolution, and cementation in the overlying reprecipitation zone occurs. This array of aragonite dissolution and reprecipitation zones potentially leads to self-organized diagenetic formation of limestone-marl rhythmites (Munnecke and Samtleben, 1996; Munnecke, 1997).

Fig. 3: Schematic diagram of the diagenesis model for rhythmic formation of calcareous nodule layers of Jenkyns (1974). (I) Dissolution of aragonite near the top of the sediment column supplies calcium and carbonate ions, which diffuse downward to sites of calcite cementation. (II) As sediment accumulates and the dissolution zone moves away from the cementation layer, the diffusion paths lengthen. (III) At a critical depth formation of new crystal seeds becomes more efficient than diffusive transport and a new layer of calcite cementation forms at a shallower depth.

Fig. 4: The cellular automaton implemented in this study comprises a rectangular matrix. Three possible states of the cells: (1) aragonitic, consisting of aragonite, calcite and clay minerals, (2) cemented, calcite cement is added to the immature sediment, and (3) non-aragonitic, either originally aragonite-free sediment consisting of calcite and clay minerals, or aragonite-depleted diagenetically mature sediment consisting of calcite and clay minerals. With each time step, the cells of a layer are moved one layer downward. As a layer passes through the aragonite dissolution zone, aragonite is dissolved from state (1) cells. For each cell from which aragonite is dissolved, a specified number of cells in the overlying cementation zone is instantly cemented.

Fig. 5: States and possible transitions of the cells used in the cellular automaton. State 1, aragonitic, consisting of aragonite and minor amounts of calcite and clay; state 2, cemented, calcite cement is added to the immature sediment of state 1 or state 3 cells;

state 3, non-aragonitic, consisting of clay and calcite. State 3 cells occur as immature sediment or are generated by dissolution of state 1 cells. State 3 cells pass the dissolution zone without modification.

Fig. 6: Example for the search of a cementation site. A 5x5 cells cementation matrix, a very low seeding probability and a six-step search for a site with well-cemented surroundings are used. 1: Random trial finds an already cemented cell and therefore fails. 2: Second trial selects a cell without cemented neighbours. Seeding fails. 3: A cell with two neighbours is picked. Search for a cell with more cemented neighbours starts (a-f). a: Three cemented neighbours, better than previous cell. b: Only one neighbour, discarded. c: Five cemented neighbour cells, better than previous cell. d: Only 4 neighbours, discarded. e: already cemented, discarded. f: already cemented, discarded. Cell picked in step "c" is finally chosen for cementation.

Fig. 7: Simulation runs of differential diagenesis by dissolution-precipitation without external trigger. Cemented receiver cells are shown in white, uncemented donor cells are black. Each panel displays a field of 1492 layers, 656 cells wide. Parameters for simulation runs are shown in Table 1. (a) Deterministic mode without random processes. The thickness of the cemented beds is determined by the distance between the dissolution layer and the cementation layer. (b) Diffuse cementation resulting from incorporation of random processes in the model. The amount of aragonite available for dissolution is limited (17%) but for each dissolved cell four other cells are cemented, resulting in an overall cementation density of 68%. Cementation takes place in a wide cementation zone. A low seeding probability results in patchy coagulation of cemented cells. The thin grey zone at top is pristine aragonite before dissolution. (c), (d) Similar conditions as in b, but with very low seeding probability resulting in larger cemented patches. A thick cementation zone (d) promotes formation of compact cement patches, while a thinner cementation zone favours dendritic growth (c). (e) Spontaneous development of cemented lenses with increasing density of dissolvable aragonite. As this increase proceeds very slowly (1% per layer) and as aragonite and cemented cells are randomly distributed, no synchronization is achieved. Therefore cementation forms diachronous layers, which may even be in antiphase (right margin). (f) While the initial conditions of a simulation run may lead to a synchronization of cemented cells that form beds (bottom), such beds will disintegrate quickly due to the lack of a further synchronization mechanism, i.e., an external trigger.

Fig. 8: Simulation runs with external trigger. Each column shows a succession of 5000 layers. Cemented receiver cells are shown in white, uncemented donor cells are black. Simulation parameters are given in Table 2. (a) A trigger with a recurrence period much longer than the period of formation of the diagenetic beds generates bundles of cemented layers alternating with thick weakly cemented layers. Note the tendency of the single beds within a bundle to become increasingly diffuse upwards, which is counteracted by the trigger pulses. Varying recurrence times of the trigger pulses generate bundles of varying

thickness. (b) Trigger with recurrence intervals shorter than in (a) generates thinner bundles. (c) Six experiments with trigger recurrence intervals shorter than the period of formation of the diagenetic beds. In each experiment the same trigger sequence is used, only the depth of the dissolution zone varies. Thicknesses of the bed-interlayer couplets that would form without the interference of a trigger are shown at the base of each column. Note the different bedding styles generated by the interaction of different diagenetic layer thicknesses with the same trigger intervals.

Fig. 9: Percentages of cemented cells in the diagenetically mature sediment (black lines) plotted with the aragonite content (shaded area) of the pristine sediment in the same layer. The units of the horizontal axes are model time steps. (a) Bundles of diagenetic beds. Same experiment as in Fig. 8a. Note the decay of amplitude, indicating the contrast between cemented beds and weakly cemented interlayers, in each bundle. Without the interaction of the trigger pulses the self-organized separation of cemented and uncemented layers would quickly give way to homogeneous cementation. (b) Phase locking of the diagenetic bedding to a trigger. Diagenetic bedding initiated in phase with the trigger (left) is quickly shifted to an antiphase position with the trigger. Resonance between trigger and diagenetic bedding consequently leads to amplification of the diagenetic amplitude, i.e., to a higher contrast between cemented and weakly cemented layers. (c) The interaction between trigger and diagenetic bedding leads to complex bedding patterns. Note that the cemented beds do not always coincide with initially aragonite-rich layers. Couplets of cemented and weakly cemented beds do not correspond to the external trigger in a one-to-one fashion. Cemented beds are shifted vertically with respect to the initially aragonite-rich layers. The number of cemented beds does not correspond to the number of the initially aragonite-rich layers.

Fig. 10: Maximum entropy periodograms of simulations with external trigger. Power spectra of the pristine aragonite content (trigger signal, thin dashed lines) and of the degree of cementation (thick lines) of the resulting diagenetically mature sediment are compared. (a) Same experiment as shown in Fig. 8b. The quasiperiodic trigger signal produces a broad peak that is also visible in the periodogram of the diagenetically mature sediment. The latter, however, shows additional power at higher frequencies that are not present in the trigger signal. These periodicities are generated by the diagenetic self-organization process. They do not carry any information about the external trigger signal. (b) Experiments shown in Fig. 8c. Quasiperiodic trigger with a mean recurrence interval of about 200 time steps interacting with diagenetic cycles of equal or longer durations. The characteristic periods of the diagenetic cycles, that is the duration a cycle would have without the influence of an external trigger signal, are given for each experiment together with the periodicity, the position of the spectral peaks (in brackets), resulting from the interaction between trigger and diagenetic cyclicity. Resonance effects shift the diagenetic cycles into two different frequency bands, with only one being close to the trigger frequency. The diagenetic cyclicity with a characteristic frequency close to the trigger frequency is strongly amplified through the interaction with the trigger signal. The higher

frequency peaks of the cement curves are harmonics of 3, 5, 7, etc. times the basic frequency.

## Table Captions

Table 1: Parameters of the simulation runs without external trigger shown in Fig. 7.

Table 2: Parameters of the simulation runs with external trigger shown in Figs. 8-10.  
Aragonite percentage: We used a trigger switching between on and off modes. Lower values given in the table are for the off mode, higher values are valid in the on mode.  
Trigger cycle range: A trigger cycle includes an "on mode" and an "off mode" period. A trigger cycle can vary in duration within the limits given in the table.



Fig. 1, Böhm et al.

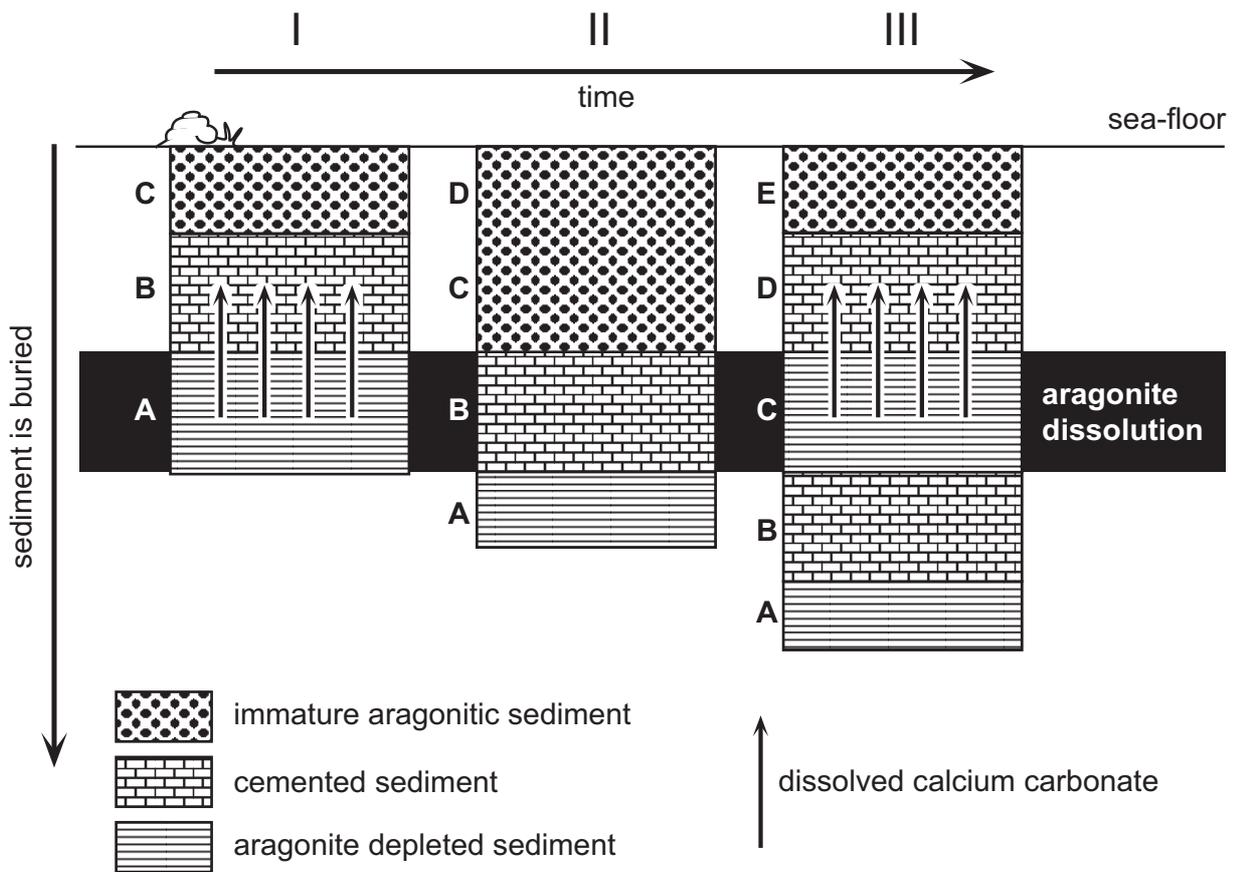


Fig. 2, Böhm et al.

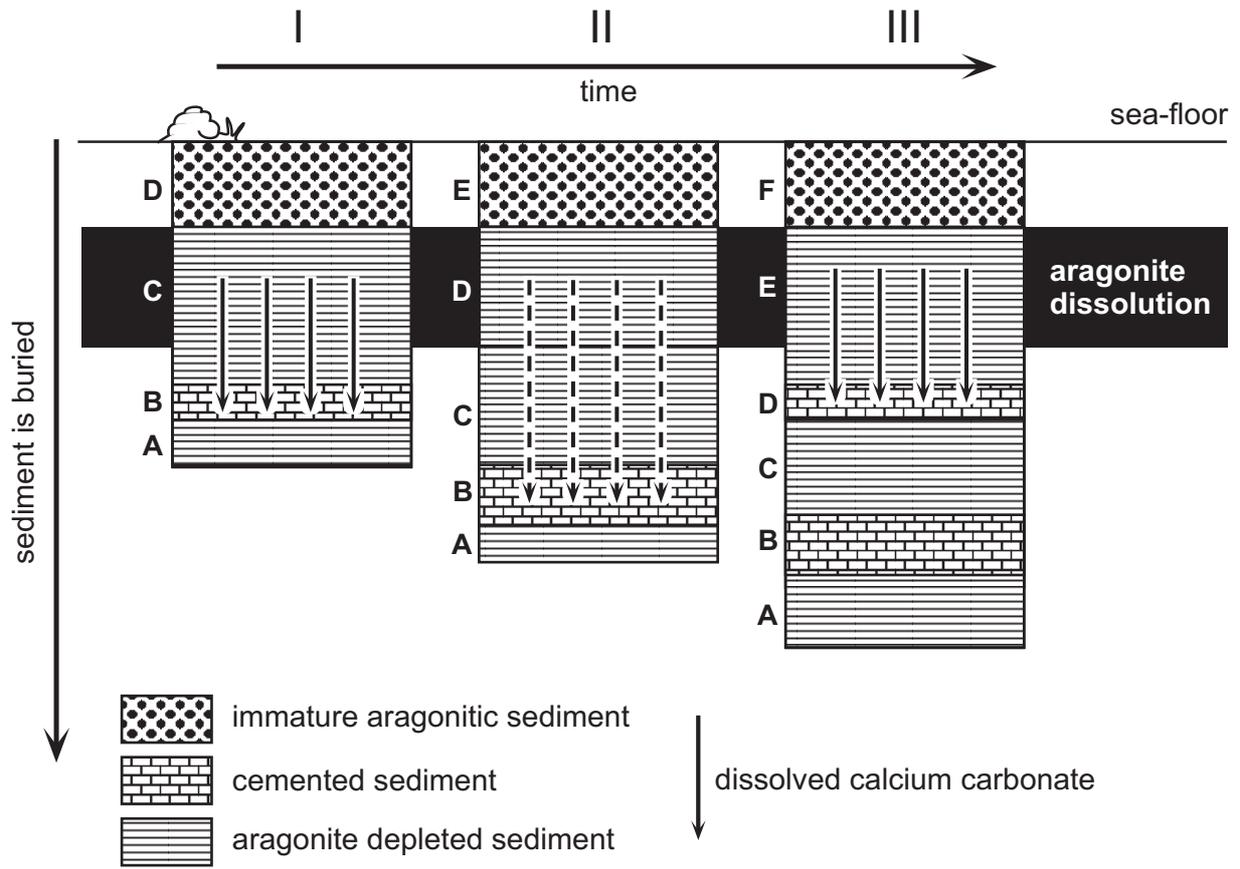


Fig. 3, Böhm et al.

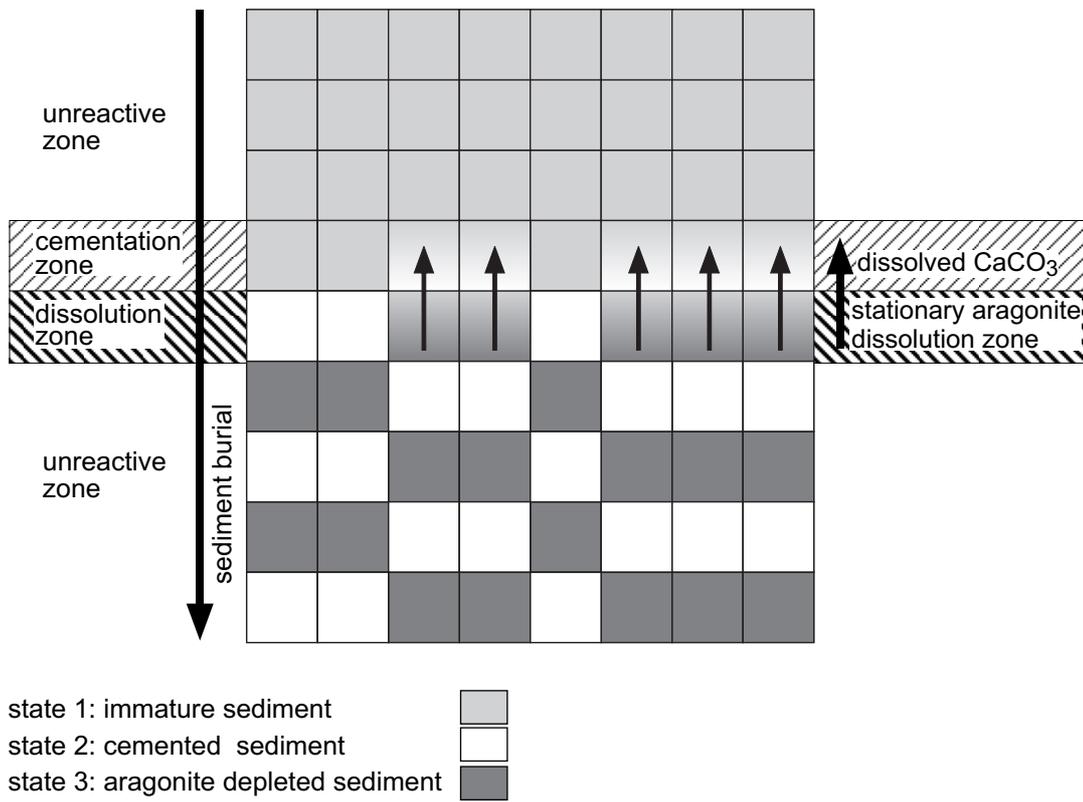


Fig. 4, Böhm et al.

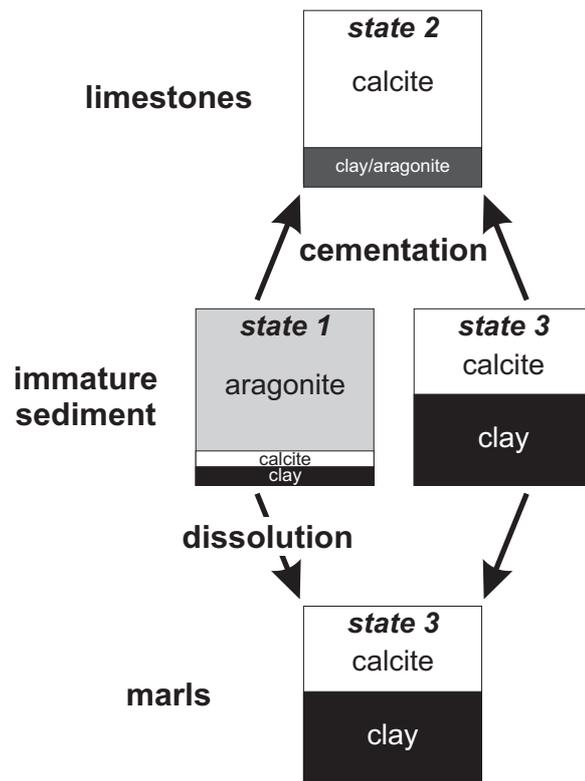


Fig. 5, Böhm et al.

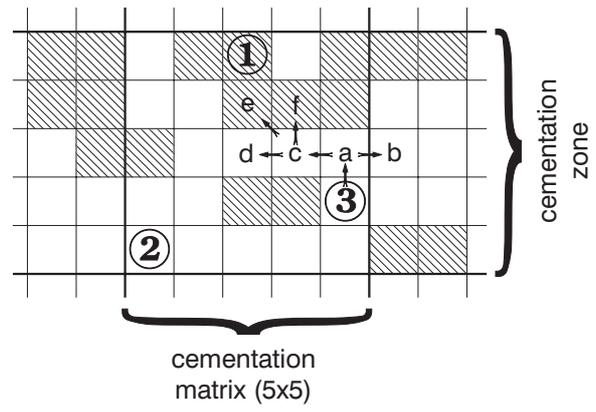


Fig. 6, Böhm et al.

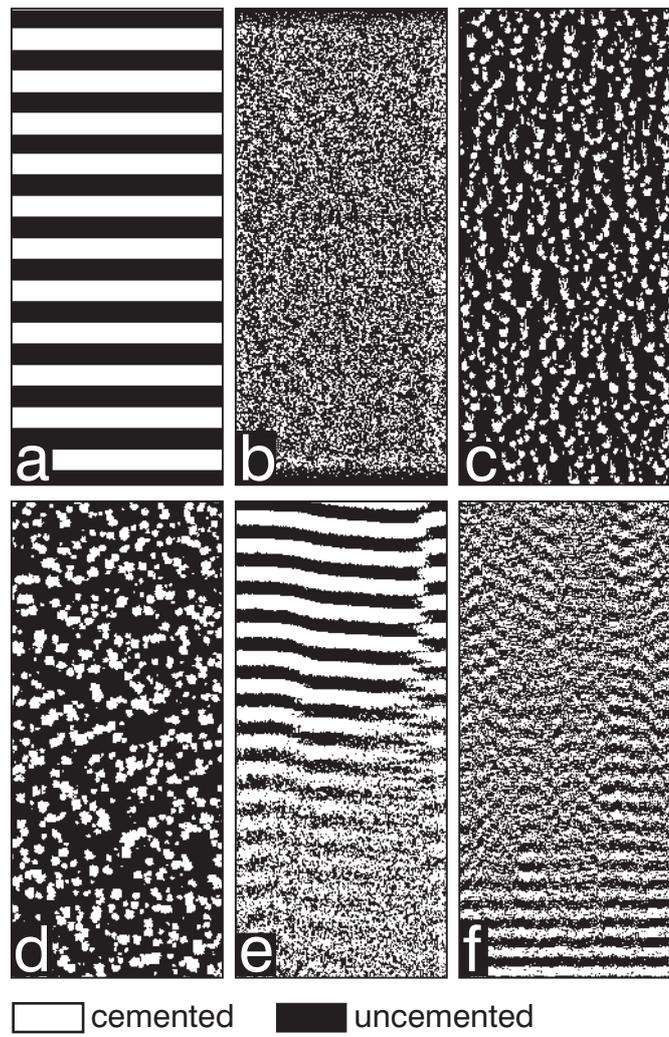


Fig. 7, Böhm et al.

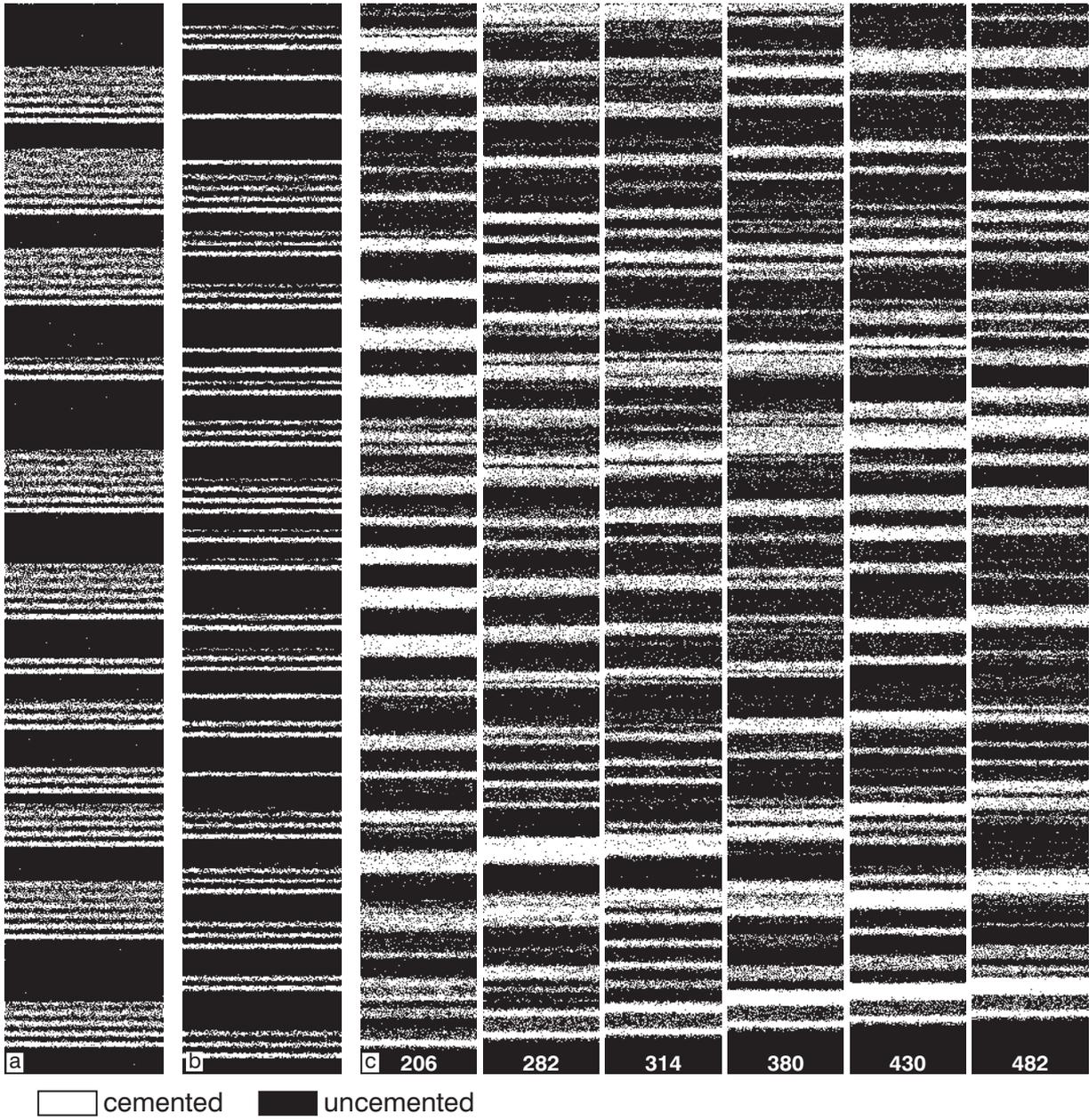


Fig. 8, Böhm et al.

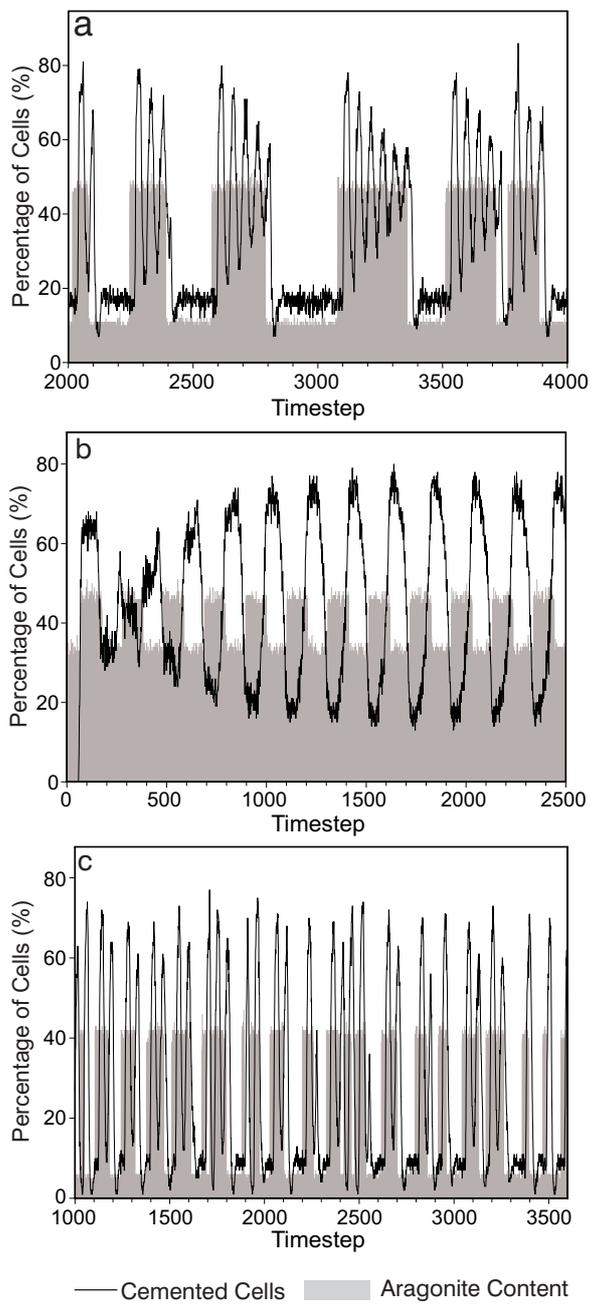


Fig. 9, Böhm et al.

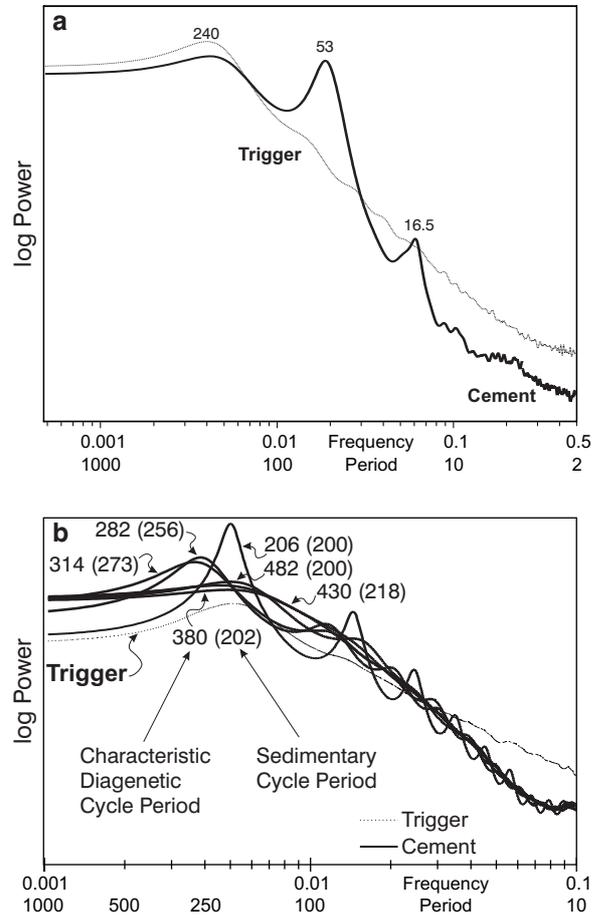


Fig. 10, Böhm et al.

	Fig. 7a	Fig. 7b	Fig. 7c	Fig. 7d	Fig. 7e	Fig. 7f
Aragonitic cells (%)	100	17	30	10	25-70	23
Cemented cells per cell dissolved	1	4	1	3	3	4
Maximum cementation density (%)	100	68	30	30	75-100	92
Seeding probability	1	0.01	0.0001	0.0001	0.01	0.1
Cementation layer thickness	1	65	30	200	7	4
Width of cementation matrix	1	171	91	59	101	13
Depth of dissolution layer top	67	67	32	1	52	32

Tab. 1, Böhm et al.

	Fig. 8a/9a	Fig. 8b/10a	Fig. 8c/10b	Fig. 9b	Fig. 9c
Aragonitic cells (%)	10/46	5/40	20/45	32/45	5/40
Trigger Cycle Range	60-660	40-400	2-270	206	4-200
Cemented cells per cell dissolved	2	2	2	2	2
Seeding probability	0.7	0.5	0.5	0.5	0.5
Cementation layer thickness	5	6	10	10	6
Width of cementation matrix	61	61	51	51	61
Depth of dissolution layer top	25	25	103-241	98	25

Tab. 2, Böhm et al.